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Abstract

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PHYSICAL CHEMISTRY

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STUDY OF THE ATMOSPHERIC CORROSION OF RADIOACTIVE SAMPLES OF "ARMCO" IRON AND STEEL-2

Until the present time, all studies on the corrosion of metals under radiation conditions have been carried out either under direct external irradiation of the metals (¹⁻³), or with the use of air ionized by radiation (⁴). We have studied the influence of an impurity of the radioactive isotope Fe-59 on the corrosion processes of "Armco" iron and carbon steel-2. This isotope has a half-life $T_{1/2} = 45.1$ days and is characterized by a complex radiation spectrum ($\beta : E_{\max} = 0.27, 0.46$ and 1.56 MeV; $\gamma : E = 0.19, 1.10$ and 1.29 MeV). The fundamental possibility of changing the character and kinetics of metal corrosion in the presence of a radioactive isotope is determined by the fact that the solid phase changes its energetic state as a result of the continuous radiation taking place (⁵).

The objects of the study were "Armco" iron and steel-2 of the following composition (content given in percent):

Composition	C	Mn	Cr	S	P	Ni	Si	Fe
"Armco" iron	0.04	0.017	traces	0.020	0.010	0.18	0.2	balance
Steel-2	0.17	0.44	0.3	0.025	0.028	traces	0.24	balance

Samples of "Armco" iron and steel-2, 0.8 mm thick, with a total surface area of each sample of 27 cm^2 , were irradiated for 48 hours with slow neutrons in a nuclear reactor at a flux of $0.87 \cdot 10^{13}$ neutrons/cm² · sec. The induced radioactivity of both the iron and the steel was 0.22 mCi/g, or 2.2 mCi/sample.

Fig. 1

Figure 1: Fig. 1

The specific activity, determined experimentally, was always somewhat higher than the calculated value, evidently due to activation of impurities.

The surface treatment of the samples in all cases was carried out with the same grade of emery paper. The samples were then washed successively with water, ether, and absolute ethyl alcohol, wiped between each washing with filter paper, and dried to constant weight. After this they were suspended on glass hooks and placed in a vessel of 1000 ml volume with a small amount of distilled water at the bottom, poured in to create a humid atmosphere. The vessel was provided with two stopcocks—at the top and at the bottom—which made it possible to carry out tests with air exchange.

The corrosion rate was studied at 100% relative humidity, i.e., in the presence of a visible moisture film on the metal. The duration of the experiments did not exceed 30 days; the temperature was maintained within the range 23–25° C. Control experiments with inactive samples were conducted in a room excluding any radiation. The amount of corrosion was calculated from data on the increase in weight of the samples, since the corrosion products formed were firmly bonded to the base metal. All work was carried out in a hot cell by means of manipulators. For weighing the samples in the hot cell, analytical damped balances with an accuracy of up to 10^{-4} g, mounted on a fixed base and adapted for remote operation, were installed.

The results of studying the corrosion of Armco iron and steel-2 are presented in Figs. 1 and 2 as curves: corrosion magnitude versus time and corrosion rate versus time. It may be concluded that, under the influence of β - and γ -radiation, the corrosion of Armco iron and steel-2 increases to a considerable degree. For specimens with a specific activity of 0.1 mCi/g, the corrosion weight gain of Armco iron over 30 days of testing exceeds that of the control specimens by 21 times, and that of steel-2 by 12 times. The corrosion rate, comparatively high on the first day, remains substantial even after 20–30 days of testing. Specimens with a specific activity of 0.2 mCi/g showed that in this case the corrosion rate of Armco iron on the 30th day of testing was 178 times higher than that of the control specimens, and that of steel-2 was 103 times higher. In the latter case, the magnitude of corrosion can also be roughly judged visually: the surface of the radioactive specimens became strongly dull during the first days of testing, and after 30 days became covered with a thick layer of dark-brown corrosion products, whereas almost the entire surface of the control specimens remained shiny, with the exception of individual dark spots.

Fig. 1. *a*—dependence of the corrosion of Armco iron active (1) and control (2) specimens on time; *b*—change with time in the corrosion rate of active (1) and control (2) specimens

Fig. 2

Figure 2: Fig. 2

Fig. 3. Dependence of corrosion of “Armco” iron and steel-2 on specific activity: 1— “Armco” iron, 2—steel-2

Figure 3: Fig. 3. Dependence of corrosion of “Armco” iron and steel-2 on specific activity: 1— “Armco” iron, 2—steel-2

It may be noted that Armco iron corrodes under irradiation conditions considerably more than steel-2, and the corrosion magnitude of both is in a definite dependence on the specific activity of the specimens (Fig. 3). The corrosion rate of Armco iron, as a result of increasing specific radioactivity, increases faster than that of steel-2. After three half-life periods of Fe-59, i.e., approximately after 4.5 months at an initial specific radioactivity of 0.22 mCi/g, the magnitude of the corrosion effect of irradiation becomes insignificant.

Fig. 2. *a*—dependence of the corrosion of steel-2 active (1) and control (2) specimens on time; *b*—change with time in the corrosion rate of active (1) and control (2) specimens

To determine the causes enhancing corrosion under the influence of radioactive radiation, experiments were carried out with grounding of the corroding specimens. In this case the possibility of anodic polarization of the surface as a result of β -radiation was assumed. In work ⁽¹⁾, the presence of a polarization effect was used to explain the considerable share of enhanced corrosion of stainless steel in a 3% NaCl solution under electron irradiation. In the case of anodic polarization of the surface, both the escape of metal ions into the electrolyte, i.e., into the moisture film, and the diffusion of oxygen to the metal surface should be facilitated. The use of grounding would eliminate polarization of the β -emitting surface. It turned out that grounding noticeably decreases the corrosion of radioactive specimens of Armco iron and steel-2; however, the radiochemical activation of the process

corrosion is far from being completely removed. Thus, anodic polarization is not the decisive factor in the enhancement of corrosion under the influence of internal radiation.

A number of authors ^(3,10) explain the intensifying action of radiation on the atmospheric corrosion of metals mainly by the formation of products of radiation-chemical transformations of water and atmospheric components, which serve as active cathodic depolarizers. However, the indicated effects were observed at higher radiation powers. In the present work the specimens had a comparatively low activity, which can hardly create an amount of radiolysis products sufficient to enhance corrosion.

Fig. 3. Dependence of corrosion of “Armco” iron and steel-2 on specific activity:

1— “Armco” iron, 2—steel-2.

X-ray diffraction study did not reveal any substantial difference in the composition of the corrosion products of radioactive and nonradioactive specimens of “Armco” iron. The corrosion products consist of γ -FeOOH with a small admixture of α -FeOOH and Fe_3O_4 .

Atmospheric corrosion with the presence of a visible moisture film on the surface has a clearly electrochemical character and in principle does not differ from the character of corrosion occurring on the surface of a metal immersed in water; therefore anodic polarization of a β -emitting surface is not the decisive cause of the increase in corrosion rate. A more probable explanation is a change in the properties of the oxide film, which may be expressed both in an increase in ionic conductivity as a result of the formation in it of defects and distortions of the crystal lattice (^{6–8}), and in the acquisition of radiation conductivity owing to the transition into the conduction band of an additional number of electrons. The latter circumstance should considerably facilitate the course of the cathodic reaction, since the corrosion film on the metal surface becomes more electrically conductive and does not hinder the escape of electrons from the metal (⁹).

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